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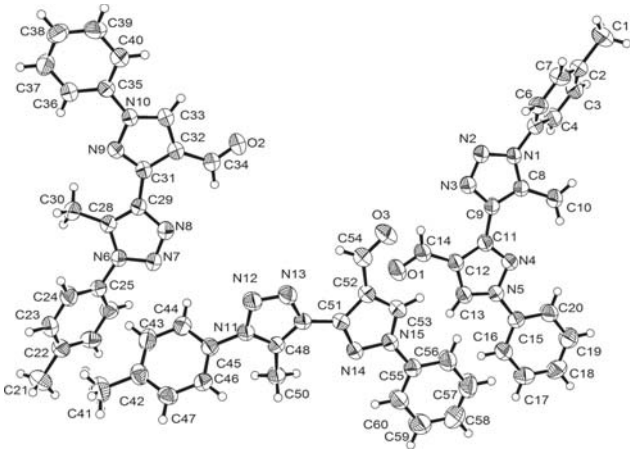
Crystal structure of 3-(5-methyl-1-*p*-tolyl-1*H*-1,2,3-triazol-4-yl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde, a rare $Z' = 3$ structure, $C_{20}H_{17}N_5O$



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Abstract

$C_{20}H_{17}N_5O$, triclinic, $P\bar{1}$ (no. 2), $a = 11.5358(7)$ Å, $b = 13.8746(9)$ Å, $c = 16.3942(10)$ Å, $\alpha = 85.958(5)°$, $\beta = 87.407(5)°$, $\gamma = 87.619(5)°$, $V = 2612.8(3)$Å^3, $Z = 6$, $R_{gt}(F) = 0.0607$, $wR_{ref}(F^2) = 0.1510$, $T = 293(2)$ K.

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The asymmetric unit of the title crystal structure, with three crystallographically independent molecules, is shown in the figure. Tables 1 and 2 contain details of the measurement method and a list of the atoms including atomic coordinates and displacement parameters.

Table 1: Data collection and handling.

Crystal:	Colorless needle
Size:	0.46 × 0.21 × 0.15 mm
Wavelength:	Mo $K\alpha$ radiation (0.71073 Å)
μ:	0.9 cm^{-1}
Diffractometer, scan mode:	SuperNova, ω-scans
$2\theta_{max}$, completeness:	60°, 82.3%
$N(hkl)_{measured}$, $N(hkl)_{unique}$, R_{int}:	23147, 12359, 0.026
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\ \sigma(I_{obs})$, 7462
$N(param)_{refined}$:	708
Programs:	CrysAlis^{PRO} [1], SHELX [2], PLATON [3], ChemDraw [4]

Source of material

3-(5-Methyl-1-*p*-tolyl-1*H*-1,2,3-triazol-4-yl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde was synthesized from treatment of 5-methyl-4-((2-phenylhydrazono)methyl)-1-*p*-tolyl-1*H*-1,2,3-triazole with phosphorus oxychloride and dimethylformamide (Vilsmeier-Haack reaction) at 0–5 °C. The reaction was left overnight at room temperature, poured onto ice-cold water and neutralized with ammonium hydroxide solution (5%). The solid obtained was filtered, dried and recrystallized from dimethylformamide to give colorless crystals of the title compound (Mp 229–230 °C) [5].

Experimental details

Non-hydrogen atoms were refined with anisotropic diaplacement parameters. All hydrogen atoms were placed in calculated positions and refined using a riding model. Methyl C—H bonds were fixed at 0.96 Å, with isotropic displacement parameters of the corresponding hydrogen atoms set to 1.5 times U_{eq}(C), and were allowed to spin about the C—C bond.

Table 2: Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å^2).

Atom	*x*	*y*	*z*	U_{iso}*/U_{eq}
C1	0.5837(3)	1.2072(2)	−0.35853(17)	0.0893(9)
H1A	0.6588	1.1886	−0.3813	0.134*
H1B	0.5863	1.2704	−0.3385	0.134*
H1C	0.5275	1.2082	−0.4001	0.134*
C2	0.5498(2)	1.13542(16)	−0.28889(14)	0.0591(6)
C3	0.4667(2)	1.15996(15)	−0.22981(14)	0.0595(6)
H3	0.4313	1.2215	−0.2334	0.071*
C4	0.43494(19)	1.09528(14)	−0.16536(13)	0.0547(5)
H4	0.3797	1.1134	−0.1256	0.066*
C5	0.48633(17)	1.00347(13)	−0.16085(12)	0.0471(5)
C6	0.56954(18)	0.97730(15)	−0.21870(13)	0.0528(5)
H6	0.6043	0.9156	−0.2154	0.063*
C7	0.6008(2)	1.04366(16)	−0.28158(14)	0.0610(6)
H7	0.6580	1.0261	−0.3202	0.073*
C8	0.34403(17)	0.90783(14)	−0.07013(12)	0.0463(4)
C9	0.36366(16)	0.83480(14)	−0.01070(11)	0.0453(4)
C10	0.23450(18)	0.95040(16)	−0.10442(14)	0.0614(6)
H10A	0.2034	0.9999	−0.0707	0.092*
H10B	0.1792	0.9008	−0.1058	0.092*
H10C	0.2503	0.9780	−0.1589	0.092*
C11	0.28006(17)	0.77584(13)	0.03711(12)	0.0464(5)
C12	0.29786(17)	0.69943(13)	0.09794(12)	0.0478(5)
C13	0.18816(18)	0.67041(14)	0.12169(12)	0.0500(5)
H13	0.1699	0.6212	0.1611	0.060*
C14	0.40469(19)	0.65853(15)	0.13092(14)	0.0606(6)
H14	0.4741	0.6764	0.1040	0.073*
C15	−0.01161(17)	0.72332(14)	0.07813(12)	0.0490(5)
C16	−0.0704(2)	0.65878(17)	0.13051(14)	0.0667(6)
H16	−0.0305	0.6182	0.1682	0.080*
C17	−0.1892(2)	0.65501(19)	0.12641(16)	0.0740(7)
H17	−0.2292	0.6110	0.1614	0.089*
C18	−0.2495(2)	0.71475(18)	0.07182(15)	0.0672(6)
H18	−0.3294	0.7107	0.0690	0.081*
C19	−0.1906(2)	0.78037(18)	0.02151(16)	0.0710(7)
H19	−0.2311	0.8224	−0.0147	0.085*
C20	−0.07219(19)	0.78484(16)	0.02390(15)	0.0637(6)
H20	−0.0327	0.8293	−0.0110	0.076*
C21	0.8783(3)	−0.01809(19)	0.85670(16)	0.0825(8)
H21A	0.8896	−0.0823	0.8387	0.124*
H21B	0.7974	−0.0055	0.8697	0.124*
H21C	0.9220	−0.0123	0.9045	0.124*
C22	0.9192(2)	0.05409(15)	0.78943(13)	0.0556(5)
C23	1.0215(2)	0.03783(16)	0.74434(14)	0.0619(6)
H23	1.0651	−0.0192	0.7552	0.074*
C24	1.06060(18)	0.10431(15)	0.68356(13)	0.0580(6)
H24	1.1296	0.0922	0.6538	0.070*
C25	0.99589(17)	0.18890(13)	0.66755(12)	0.0459(5)
C26	0.89352(18)	0.20651(14)	0.71080(13)	0.0523(5)
H26	0.8500	0.2635	0.7000	0.063*
C27	0.85592(19)	0.13852(15)	0.77060(13)	0.0571(5)
H27	0.7857	0.1501	0.7991	0.069*
C28	1.13904(17)	0.29433(13)	0.58506(12)	0.0459(4)
C29	1.12698(16)	0.34506(13)	0.51064(12)	0.0444(4)
C30	1.23670(19)	0.28372(16)	0.64173(13)	0.0607(6)
H30A	1.2062	0.2798	0.6973	0.091*
H30B	1.2847	0.3387	0.6328	0.091*
H30C	1.2821	0.2259	0.6316	0.091*
C31	1.21337(17)	0.39641(13)	0.45809(12)	0.0444(4)
C32	1.19619(17)	0.46291(13)	0.38921(12)	0.0478(5)
C33	1.30661(17)	0.48380(13)	0.35982(12)	0.0491(5)
H33	1.3257	0.5253	0.3146	0.059*
C34	1.08854(19)	0.50342(15)	0.35610(14)	0.0598(6)
H34	1.0193	0.4865	0.3837	0.072*
C35	1.50596(17)	0.42941(12)	0.40207(12)	0.0451(4)
C36	1.56754(18)	0.38670(14)	0.46653(13)	0.0534(5)
H36	1.5283	0.3636	0.5143	0.064*
C37	1.6872(2)	0.37856(16)	0.45981(15)	0.0621(6)
H37	1.7285	0.3496	0.5030	0.075*
C38	1.7455(2)	0.41297(16)	0.38959(16)	0.0653(6)
H38	1.8260	0.4066	0.3849	0.078*
C39	1.6842(2)	0.45682(16)	0.32626(15)	0.0658(6)
H39	1.7239	0.4809	0.2791	0.079*
C40	1.56400(19)	0.46557(15)	0.33182(13)	0.0553(5)
H40	1.5231	0.4954	0.2888	0.066*
C41	0.8478(2)	−0.11266(18)	0.59993(15)	0.0736(7)
H41A[a]	0.8055	−0.1709	0.6011	0.110*
H41B[a]	0.8357	−0.0840	0.6514	0.110*
H41C[a]	0.9290	−0.1276	0.5905	0.110*
H41D[a]	0.9080	−0.0841	0.6275	0.110*
H41E[a]	0.8778	−0.1710	0.5772	0.110*
H41F[a]	0.7844	−0.1274	0.6382	0.110*
C42	0.80536(18)	−0.04255(15)	0.53197(12)	0.0531(5)
C43	0.8520(2)	0.04740(16)	0.51683(14)	0.0636(6)
H43	0.9099	0.0653	0.5497	0.076*
C44	0.81505(18)	0.11137(15)	0.45443(14)	0.0574(5)
H44	0.8476	0.1716	0.4455	0.069*
C45	0.72963(16)	0.08518(13)	0.40557(12)	0.0450(4)
C46	0.68009(17)	−0.00362(14)	0.41920(13)	0.0513(5)
H46	0.6217	−0.0209	0.3864	0.062*
C47	0.71846(18)	−0.06680(14)	0.48248(13)	0.0526(5)
H47	0.6852	−0.1267	0.4919	0.063*
C48	0.58588(16)	0.18712(13)	0.32257(12)	0.0429(4)
C49	0.60166(16)	0.23997(13)	0.24931(12)	0.0448(4)
C50	0.48398(17)	0.17299(14)	0.38008(12)	0.0514(5)
H50A	0.4495	0.1130	0.3712	0.077*
H50B	0.4280	0.2254	0.3709	0.077*
H50C	0.5085	0.1714	0.4354	0.077*
C51	0.51597(17)	0.29574(13)	0.20050(12)	0.0453(4)
C52	0.53391(18)	0.36505(14)	0.13362(12)	0.0487(5)
C53	0.42401(19)	0.39389(14)	0.11154(13)	0.0525(5)
H53	0.4052	0.4390	0.0690	0.063*
C54	0.6414(2)	0.40189(16)	0.09763(14)	0.0646(6)
H54	0.7103	0.3749	0.1180	0.078*
C55	0.22398(18)	0.35042(14)	0.16356(13)	0.0515(5)
C56	0.1655(2)	0.40945(16)	0.10694(14)	0.0631(6)
H56	0.2061	0.4479	0.0676	0.076*
C57	0.0459(2)	0.4108(2)	0.10937(17)	0.0781(7)
H57	0.0059	0.4502	0.0710	0.094*
C58	−0.0146(2)	0.3555(2)	0.16704(19)	0.0864(8)
H58	−0.0953	0.3571	0.1679	0.104*
C59	0.0438(2)	0.2976(2)	0.2237(2)	0.0946(10)

Table 2 (continued)

Atom	x	y	z	U_{iso}*/U_{eq}
H59	0.0028	0.2598	0.2633	0.114*
C60	0.1629(2)	0.29523(18)	0.22220(17)	0.0786(8)
H60	0.2025	0.2561	0.2610	0.094*
N1	0.45217(13)	0.93438(11)	−0.09626(9)	0.0467(4)
N2	0.53532(14)	0.88023(12)	−0.05527(10)	0.0543(4)
N3	0.48085(14)	0.82014(12)	−0.00338(10)	0.0541(4)
N4	0.16724(14)	0.79213(12)	0.02524(10)	0.0511(4)
N5	0.11219(14)	0.72591(11)	0.07767(10)	0.0487(4)
N6	1.03332(14)	0.25626(11)	0.60200(10)	0.0463(4)
N7	0.95884(14)	0.28316(12)	0.54184(11)	0.0545(4)
N8	1.01618(15)	0.33728(12)	0.48647(11)	0.0535(4)
N9	1.32598(14)	0.37813(11)	0.46925(10)	0.0466(4)
N10	1.38207(14)	0.43328(11)	0.40828(9)	0.0451(4)
N11	0.69354(13)	0.14981(11)	0.33874(10)	0.0464(4)
N12	0.77293(15)	0.17919(13)	0.27893(11)	0.0582(5)
N13	0.71603(15)	0.23375(12)	0.22477(11)	0.0572(5)
N14	0.40405(14)	0.28389(11)	0.21795(10)	0.0497(4)
N15	0.34801(14)	0.34560(11)	0.16206(10)	0.0492(4)
O1	0.41041(14)	0.60292(12)	0.19079(11)	0.0797(5)
O2	1.08280(14)	0.55765(11)	0.29511(10)	0.0737(5)
O3	0.64867(16)	0.46490(13)	0.04334(11)	0.0873(6)

[a]Occupancy: 0.5.

Aromatic C—H distances were set to 0.93 Å and U_{iso} parameters of aromatic hydrogen atoms were set to 1.2 times the U_{eq} of the atoms to which they are bonded. The methyl group C41 shows a disorder of its hydrogen atoms (*cf*. the figure).

Comment

Compounds containing a pyrazole ring system [6–10] exhibit a wide range of biological applications. Hence, it has been shown that many derivatives show antimicrobial, fungicidal, anticancer and antioxidant activities [11–14].

The asymmetric unit comprises three crystallographically independent molecules of $C_{20}H_{17}N_5O$. Molecular conformation in the three molecules is very similar with twist angles between the triazole, pyrazole and phenyl rings ranging from 2.40(14) to 18.71(11)° (*cf*. the figure). The twist between the triazole and methylbenzene groups is greater in all three molecules, being in the range 47.53(8)–58.30(8)°. In the crystal structure, the molecules stack with planes roughly parallel to (011). All bond lengths and angles are in the expected ranges.

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